

Comment on ‘Phase-separated states in antiferromagnetic semiconductors with a polarizable lattice’

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Abstract

The ‘proof’ of the possibility of the slab or stripe phase separation in polar semiconductors, proposed recently by E. L. Nagaev (Phys. Rev. **B64** (2001) 014401) is shown to be erroneous. The fundamental error originates in the double-counting of the electron-phonon interaction with optical phonons.

PACS numbers:74.20.-z,74.65.+n,74.60.Mj

In a recent publication [1] Nagaev claims that he 'managed to prove' the possibility of charge segregation in antiferromagnetic semiconductors with a polarizable lattice. The author neglects the bulk of earlier results including the pioneering works by Pekar [2], Vinetskii and Gitterman [3] and more recent studies [4–14], where the problem of bound states of a few and many polarons was properly addressed in both large and small polaron cases. The conclusions of Ref. [1] are in diametric contradiction with those and many other studies. Here I show that Ref. [1] is wrong on an elementary level.

A possibility of pairing of two large polarons was considered by Pekar half a century ago [2]. He found that a large bipolaron does not exist independently of the crystal parameters, ϵ_∞ and ϵ_0 . Physically, one can reach this conclusion by scaling arguments [4]. The long range interaction with optical phonons is Coulomb like at large distances. Then the total energy of a large polaron, which must be minimised is

$$E_p(r) = \frac{\pi^2 \hbar^2}{2mr^2} - \frac{e^2}{2\epsilon^* r}. \quad (1)$$

The first term in Eq.(1) is the kinetic energy of a particle confined in a sphere of radius r , while the second term is the potential energy due to lattice polarisation ($1/\epsilon^* = \epsilon_\infty^{-1} - \epsilon_0^{-1}$). Minimising Eq.(1) with respect to r one obtains the polaron binding energy

$$E_p = -\frac{1}{4\pi^2} \alpha^2 \hbar \omega, \quad (2)$$

where α is the Fröhlich coupling constant with the optical phonons of a frequency ω . For a state of two carriers sharing the same orbital within a common potential well the corresponding functional is

$$E_b(r) = 2\frac{\pi^2 \hbar^2}{2mr^2} - 4\frac{e^2}{2\epsilon^* r} + \frac{e^2}{\epsilon_\infty r}, \quad (3)$$

where the first term is twice the polaron kinetic energy, the second term is four times the corresponding term for a polaron because the polarisation is twice as large as that for a polaron, and the last term describes the (bare) Coulomb repulsion between two carriers.

The large bipolaron is energetically stable with respect to dissociation into two separate large polarons if the binding energy is positive,

$$\Delta \equiv 2E_p - E_b > 0. \quad (4)$$

This however is not the case because [4]

$$\Delta = 2|E_p| \left[\left(1 - \frac{\epsilon_\infty}{\epsilon_0 - \epsilon_\infty} \right)^2 - 1 \right] < 0. \quad (5)$$

While a large portion of the Coulomb repulsion is nullified by the Fröhlich electron-phonon interaction the latter interaction alone remains insufficient to produce a bound state. That is because $\epsilon_0 > \epsilon_\infty$. Approaching the problem from the weak coupling limit Takada [5] and Khomskii [6] reached the same conclusion. However, in a strongly polarisable lattice with $\epsilon_0/\epsilon_\infty \gg 1$ the absolute value of the bipolaron binding energy $|\Delta|$, Eq.(5) is very small

$$\frac{\Delta}{2E_p} \simeq 2 \frac{\epsilon_\infty}{\epsilon_0} \ll 1. \quad (6)$$

Therefore one can expect that the wave functions of two polarons strongly overlap, so that the quantum *exchange* interaction can stabilise Pekar's bipolaron even without any other additional attraction but the Fröhlich one. This was first realised by Vinetskii and Gitterman [3], and confirmed in a number of other comprehensive studies [7–9]. The most reliable path-integral approach [9] proved that large bipolarons might be formed if the ratio of the static and high-frequency dielectric constants is very large, $\epsilon_0/\epsilon_\infty \gg 1$. The formation of three-polaron bound states, as well as many-polaronic droplets, slabs, stripes and strings was shown to be impossible with the long-range Fröhlich or zero-range Holstein interactions [10,13,14].

Surprisingly, the author of Ref. [1] reaches the opposite conclusion. He introduces the Hamiltonian (Eq.(1) of Ref. [1]) with the Fröhlich interaction, H_{sp} (Eq.(4) of Ref. [1]) and the *renormalised* Coulomb interaction, H_c , defined with the *static* dielectric constant, $H_c \propto 1/\epsilon_0$ (Eq. (11) of Ref. [1]). This is in disagreement with all textbooks, and previous studies, where the Coulomb interaction is defined with the high-frequency dielectric constant, $H_c \propto 1/\epsilon_\infty$. As a result, the author finds a 'polaron instability' occurring at $\epsilon_0 = 2\epsilon_\infty$ (Eq.(24) of Ref. [1]). Analysing the problem of phase-separated states he further claims that 'if one wishes to take into account the polaronic contribution, one should replace ϵ_0^{-1} (in the

Coulomb term) by $\kappa^{-1} = \epsilon_0^{-1} - 2\epsilon_\infty^{-1}$, (Eq.(27) of Ref. [1]). This substitution leads him to a conclusion that a charge segregation appears in manganites with the values of $\epsilon_0 = 5$ and $\epsilon_\infty = 3.4$. The author fails to understand that an enhanced value of the static dielectric constant in polar semiconductors is mainly due to the Fröhlich interaction itself. By taking the Coulomb repulsion in his Hamiltonian with the *static* dielectric constant, he already accounts for the attractive interaction between carriers mediated by the optical phonons. Introducing H_{sp} as an independent term in the Hamiltonian leads to the obvious double-counting of the electron-phonon interaction with such a choice of the Coulomb repulsion. Instead of replacing ϵ_0 in the incorrect H_c with incorrect κ , one could rather replace the high-frequency dielectric constant, ϵ_∞ , in the *correct* $H_c \propto 1/\epsilon_\infty$ with ϵ_0 to account for the Fröhlich interaction. Different from κ in Eq.(27) of Ref. [1], the static dielectric constant is always positive. Hence, the effective polaron-polaron interaction is always repulsive at large distances, so that there is no phase separation of the Fröhlich polarons [14]. When an additional finite-range interaction with the deformation potential or with the antiferromagnetic fluctuations is introduced, E_{att} , only short-length stripes are theoretically possible [12,14]. The number of polarons bound in the stripe is estimated as [14]

$$N = \exp \left(\frac{\epsilon_0 a E_{att} \delta \omega}{e^2 \omega} - 2.31 \right), \quad (7)$$

where ω is the characteristic frequency of acoustic phonons or magnons responsible for the finite-range attraction, $\delta \omega$ its maximum dispersion, and a is the lattice constant. Then, with Nagaev's value of $\epsilon_0 = 5$, and $a = 3.8 \text{ \AA}$, there is no charge segregation at all because $N < 2$ for any realistic $E_{att} \leq 1 \text{ eV}$. Hence, it is more probable that the polarons in oxides remain in a charge-homogeneous state. Indeed, there is a growing understanding that the coexisting phases in manganites must have nearly the same charge densities [15–17]. Ref. [1] is wrong from beginning to end on a rather elementary level.

I would like to thank Victor Kabanov for calling my attention to Ref. [1] and the EPSRC for partial support (grant R46977).

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